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Thermo-Mechanical Properties of a Metal-filled Polymer Composite for Fused Deposition Modelling Applications

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Abstract: Thermo-mechanical properties of a new metal-polymer composite consisting of an FDM-grade acrylonitrile butadiene styrene (ABS) containing 10% fine iron particles by volume have been investigated experimentally. Thermal properties tested include glass transition temperature using Dynamic Thermal Analysis (DTA) and Heat Capacity using Differential Scanning Calorimetry (DSC). The tensile strength and dynamic mechanical properties have also been tested. It has been shown that the addition of 10% iron powder improves thermal properties and storage modulus of the FDM-grade ABS resulting in more thermally stable prototypes producible on Stratasys FDM300 machine, while tensile strength drops significantly. The feedstock filaments of this composite have been successfully produced and used in the Fused Deposition Modelling (FDM) rapid prototyping machine.

Keywords: Fused Deposition Modelling (FDM); Rapid prototypes; Composite materials; Thermal properties; tensile strength 1 Introduction

1 Introduction

The Fused Deposition Modeling (FDM) rapid prototyping systems, developed by Stratasys Inc., can fabricate parts in a range of materials including elastomers, ABS and investment casting wax with the layer by layer deposition of extruded material through a nozzle using feedstock filaments from a spool. Most of the parts fabricated in these materials can be used for design verification, form and fit checking and patterns for casting processes and medical application. New materials for FDM process are needed to increase its application domain especially in rapid tooling and rapid manufacturing areas. The basic principle of operation of the FDM process, as shown in Figure 1 [1], offers great potential for a range of other materials including metals and composites to be developed and used in the FDM process as long as the new material can be produced in feedstock filament form of required size, strength and properties.

Research has been going on in universities and research institutions around the world to increase the applications of FDM and to improve the FDM process [2]. Work has also been in progress in some organizations to develop new metallic and ceramic materials for rapid fabrication of functional components by FDM with higher mechanical properties. Researches at Rutgers University in the US have carried out considerable work in the development of fused deposition of metals [3]. They create such components on the FDM using metal powders mixed with organic binder system. The properties of the mixed feedstock filament meet the flexibility, stiffness, and viscosity required for successful FDM processing. But the fabricated green parts need to undergo further processing to remove the organic binder and are subjected to sintering to achieve densification. Sintered part may be infiltrated with other type of metal materials.

This paper presents the development and characterization of a iron-filled FDM-grade ABS composite material for direct FDM processing being carried out at Swinburne University of Technology without the need of binder removal or infiltration process. The aim is to develop the new composite with desirable thermo-mechanical properties for producing rapid tooling for injection molding application.

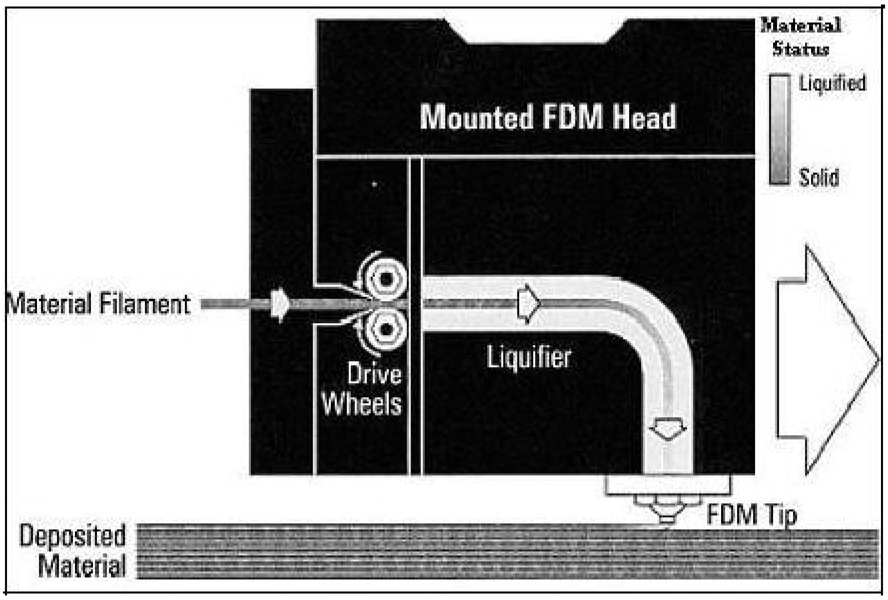

Fig 1 Schematic of Stratasys FDM Process [1]

2 Experimental Procedures

2.1 Preparation of Iron/ABS Composite

To develop the new metal-polymer composite, a mixture of 10% iron powder and 90% ABS powder by volume were selected with the aim of producing appropriate feed stock filament for FDM processing. The main reasons for selection of iron powder as short fiber fillers were its reasonably good mechanical and thermal properties as well as its capabilities of mixing and surface bonding with polymers. Iron powder was purchased from Sigma-Aldrich in Australia. The purity of powder was 99.7% with average particle size of $45\ \mu m$. The specific gravity of iron powder was $7.88\ gr/cm^3$ and the shape of the iron particles was spherical.

The polymer used was P400-grade acrylonitrile butadiene styrene (ABS) supplied by the Stratasys Inc. This ABS is the FDM-grade polymer recommended by the Stratasys for use in fabrication of protopyes on their FDM3000 machine. The specific gravity of ABS was $1.05\ gr/cm^3$.

To produce ABS particles, sufficient amount of P400 filmanet was first pelletized on a mechanical chopper. Then the ABS pellets were ground to fine pwders using the cryogenic grinding technique. The machine used for this purpose was a SORVALL OMNI high speed grinder operating at sub-zero temperatures. During this process, the ABS pellets were frozen by the surrounding liquid nitrogen which resulted in lower molecular energy of the pellets. Simultaneously, high speed rotation of stainless steel blades within the chamber containing the ABS pellets could easily break them below the glass-transition temperature. This process does not damage or alter chemical compostion of material making it a very efficient polymer powder production technique**.** The grinding equipment was provided by CSIRO, Australia.

In order to achieve a homogeneous mixture with higher packing factor when mixed with iron particles an ABS/Iron particle size ratio of 10 to 1 was needed [4]. Therefore, the ABS pellets were ground to a particle size of approximately $450-500\ \mu m$. To get the same size for ABS particles, grinding process was done in three time-interval of five minutes between which the particles were sieved to the appropriate size. This helped to screen the particles of different size range than $450-500\ \mu m$. In the next step, ABS and iron powders were mixed and placed in a ball-mill mixer to achieve maximum possible homogenous-distribution of iron powder in ABS matrix. At the end, a very small percentage by weight of a surfactant was added to the mixture. According to the previous studies carried out at Swinburne involving iron and nylon [5], addition of surfactant increased homogeneous dispersion of metal particles in polymer matrix. The surfactant powder is coated on the iron particles reducing the high free energy surfaces of the iron fillers. The coated iron particles gave good link to lower free energy surfaces of polymer particles.

2.2 Fabrication of FDM filament and test samples

The filament used in FDM process needs to be of a specific size, strength and the properties. A single screw extruder was selected to fabricate the filaments from the composite mixture. Due to die swell phenomenon during the extrusion process of polymeric materials, there is always slight difference between dimensions of the extrusion die and those of the extrudate. To minimize this difference and achieve a consistent diameter on the extrudate in such a way that the produced filament could be fed into the FDM machine smoothly, different variables including screw speed, pressure and temperature were examined and selected until an optimum dimension (diameters of 1.75-1.80 mm) for the filament was achieved.

In order to create a part on the FDM system using the new composite material, a certain amount of this composite is required to create the filaments for FDM machine. This amount of required composite material must have exact amount of its constituent elements, which include ABS, iron, and surfactant. The amount of each of these elements will depend upon the volume of the filaments required for FDM processing. In this experiment, the exact amount of constituents was determined by considering the CAD model volume. The weight of the composite was calculated by the following relationship:

$$Wc = \frac{W_{Fe} + W_{ABS}}{(1 - W_s\%)} \qquad (1)$$

where W_c, W_{Fe}, W_{ABS} are the weight of composite, iron, ABS respectively, and W_s is the weight percentage of surfactant used. Figure 2 shows the final filament produced by this process. Figure 3 shows samples of parts produced from the new composite material on the FDM3000 system.

Fig 2 FDM filament produced from Iron/ABS composite material

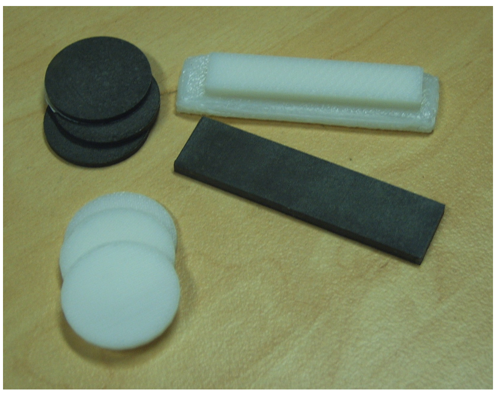

Fig 3 Test samples produced on FDM3000 from the new Iron/ABS composite (black colour) and unfilled ABS (white colour)

3 Determination Of Thermo-Mechanical Properties

Dynamic mechanical analysis was conducted on a Multi-Frequency-Dual Cantilever DMA Instrument which is an ideal experiment for rapidly screening and comparing the mechanical properties of the materials such as Storage Modulus and Loss Modulus as well as glass transition temperature. In this method, the material is heated at a constant rate and deformed (oscillated) at a constant amplitude (strain) and frequency. The test mode applied was single frequency one with amplitude of $15\ mm$ with a temperature ramp of 5 ^oC/min upto 150 ^oC. Data sampling interval was 2 sec/pt.

Heat capacity and heat flow were measured using standard Differential Scanning Calorimetry modulated at +/- 5 ^oC at every 40 seconds with temperature ramp of 3 ^oC/min up to 150 ^oC. To measure maximum load and elongation at break point, standard tensile test was conducted on a Zwick/Z010 Instrument at a speed of 50 mm/min. At least three samples were prepared for each test. The average values have been plotted.

4 Results And Discussion

Storage Modulus and Loss Modulus of the new composite material have been compared to those of the virgin material. Figure 4 shows the comparison of the graphs of dynamic mechanical properties of iron-powder filled ABS composite and unfilled ABS. The solid line shows the properties of filled ABS while broken lines represent properties of virgin ABS. As it can be seen from the graph, introduction of 10% iron powder has increased the storage modulus by nearly 40%. Glass transition temperature represented on Tan Delta curve has shifted by 7 degrees Celsius. By further increase of glass transition temperature, softening point of the new composite material will be higher, which gives the promise of using the new material as die or insert material for injection molding of polymers and plastics with lower softening point.

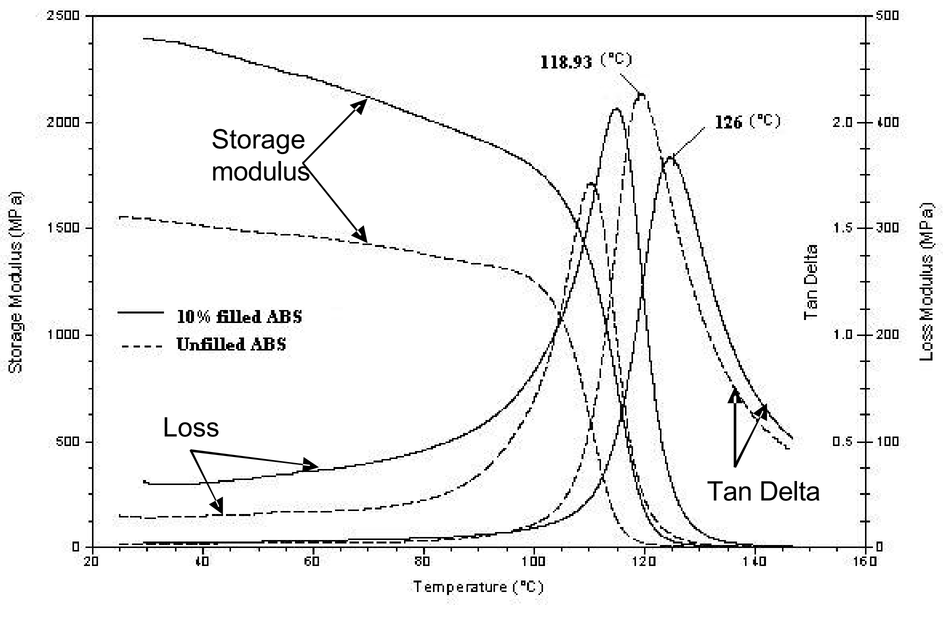

Fig 4 Comparison of dynamic mechanical properties of virgin acrylonitrile butadiene styrene (ABS) and 10 % iron-powder filled ABS

Figure 5 shows the graphs of heat capacity variation with temperature for virgin ABS and composite materials with 10% iron and 20% iron powder. It shows that 10% Fe decreases heat capacity of the unfilled ABS. Further addition of iron powder confirms the same trend of reduction in heat capacity which on the other hand means the thermal conductivity increases by approximately the same percentage. Increase of thermal conductivity is another advantage of the new material by which much more thermally stable prototypes can be produced on FDM machine making them dimensionally more accurate and reliable for reducing the cooling cycle time when employed as material for injection molding tools.

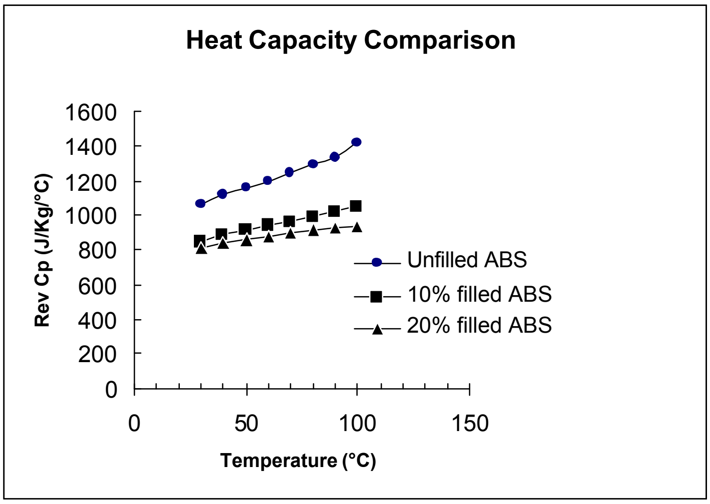

Fig 5 Heat capacity rated for metal-polymer matrix composite

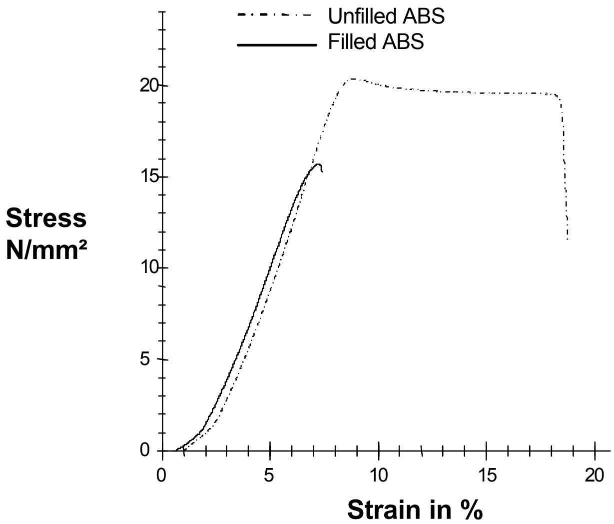

Fig 6. The effect of Fe particle on tensile strength of ABS

Figure 6 shows the effect of iron powder on the tensile strength and elongation. As it can be seen the behavior of iron filled ABS is of characteristics of a brittle and hard material with much lower elongation. Table 1 shows the comparison of actual values of maximum elongation at maximum load and at break point along with the values of maximum load and load at break for the unfilled ABS and the iron filled ABS. Although there is only slight difference on elongation at maximum load, it becomes significant at break point. Tensile strength drops significantly as a result of addition of 10% volume percent of iron powder. It should be noted that the current composition of the new material does not contain any additive which could possibly promote better bonding between iron particles and polymer matrix. According to a work reported by Bigg [6] there are factors that are responsible for integrity and long-term durability of metal-polymer bonds. These factors include morphology of the surface oxide on the metal and environmental stability of the same oxide films. These issues are under investigation at Swinburne for development of stronger metal-polymer bonds where along side the improvement of thermal properties, higher mechanical strength can also be achieved for rapid tooling applications.

Table 1 Comparison of Elongation Test values at maximum load and at break

Sample	Fmax N	F_{Break} N	σBreak mm	σFmax mm
Unfilled ABS	709.3	690.7	2.06	2.02
Filled ABS	609.9	344.9	5.32	2.37

5 Conclusions

A new composite material with iron filled particle in ABS polymer has been successfully developed for direct application in Fused Deposition Modeling rapid prototyping process. The flexible filaments of the new material have been successfully produced and processed in the existing FDM3000 machine to produce sample parts. Material characterization of the new material has been investigated. Comparison of the dynamic mechanical test values and heat capacity values for the virgin ABS and the new composite materials show great promise for application of the new material in developing tooling inserts and dies directly on the FDM system.

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